

### **REMARKS/ARGUMENTS**

Claims 1, 3, 5-15, 18-25, 29-31 and 36-39 remain in this application. Claims 2, 4, 16, 17, 26-28 have been canceled and claims 32-35 have been withdrawn. Claims 1, 18, 25, 36 and 39 are being currently amended. Reconsideration of this application in view of these amendments and the following remarks are respectfully requested.

Claims 1-15, 18-21 and 24 of this application stand rejected under 35 U.S.C. §103 as unpatentable over Soni and Crynes, "A comparison of the Hydrodesulfurization and Hydrodenitrogenation Activities of Monolith Alumina Impregnated with Cobalt and Molybdenum and a Commercial Catalyst", *ACS Symp. Ser.*, 156 (1981), 208-224 (Soni), taken in view of Irandoost and Gahne, "Competitive hydrodesulfurization and hydrogenation in a Monolithic Reactor", *AIChE Journal*, 36(5), 746-752 (1990) (Irandoost). Soni was cited to show heteroatom removal from a liquid hydrocarbon stream using a honeycomb catalyst at temperatures and pressures in the Applicants' ranges, while Irandoost was cited to show hydrocarbon treatments at higher liquid linear velocities.

This rejection is respectfully traversed for the reason that the process modifications disclosed and claimed by the Applicants produce superior results that are not predictable, and in fact entirely unexpected, from the results obtained in accordance with the cited references and other prior experience in the hydrotreating field.

The processing conditions and resulting conversion results obtained in accordance with the disclosure of Soni are reported as Prior Art results in Table 3 of the present application. The contrast between those results and the conditions and results attributable to the use of the claimed invention are evident.

Essentially, Soni discloses the hydroprocessing of a hydrocarbon (anthracene) liquid through a honeycomb catalyst at a reactor temperature of 371°C, and a reactor pressure of 103 bar, reaching a sulfur removal level of only about 76% at a low liquid hourly space velocity (LHSV) of 0.65 hr<sup>-1</sup> and a very low calculated liquid linear velocity of about 0.01 cm/sec. These are not commercially viable processing conditions.

The contrasting data in Table 3 for the Applicants' process establishes that sulfur removal rates in excess of 80% are routinely achievable at the same temperatures, and at

even lower pressures, at space velocities (LHSV) in excess of  $14\text{hr}^{-1}$  and liquid linear velocities in excess of 0.34 cm/sec. As the Applicants have pointed out at page 13, lines 9-18 of the specification:

...the HDS conversion data for the comparative example [\*Soni and Crines] set out in Table 3 above indicates relatively poor performance for the conversion of a raw anthracene feed utilizing the catalyst characterized in that report. Even at similar reaction temperatures, the prior art monolithic reactor achieved a lower HDS conversion rate than any of the monoliths operated under the reaction conditions found effective for efficient HDS conversion in accordance with the invention. This is particularly surprising in view of the fact that the comparative example was carried out at a higher reactor pressure and at a lower liquid hourly space velocity than employed in accordance with the inventive examples—differences that typically would produce higher conversion rates. (emphasis added)

Thus, what the Applicants have contributed to the art is the entirely unexpected finding that commercially useful levels of heteroatom removal can be achieved with appropriate honeycomb catalysts if higher space velocities and liquid linear flow velocities are used in processing. Certainly there is nothing whatever in Soni to suggest this result.

In recognition of the fact that Soni fails to teach or suggest the Applicants' commercially viable process, the Examiner cites Irandoost to suggest sulfur removal processing at higher liquid flow rates. At least two factors weigh against the Examiner's conclusion that the use of higher (Irandoost) flow rates in the method of Soni would be obvious. First, it is clear from the data presented by Soni in Figs. 5 and 6 of the reference that one-pass sulfur and nitrogen removal rates drop drastically in the Soni reactor at higher liquid processing rates (shorter hourly space times). Certainly there could be no expectation whatever of achieving higher one-pass sulfur removal rates at higher liquid processing rates in light of this specific experimental data.

Secondly, the reactor of Irandoost is not a one-pass reactor for piloting a commercial process but rather a laboratory recycle reactor for measuring catalyst activity parameters wherein high levels of sulfur removal from a model hydrocarbon feed are achieved only after multiple passes through the catalyst bed. In particular, the data in Fig. 6 indicates a slow reduction in thiophene concentration in the model feed over a

period of hundreds of minutes, with an 80% reduction in concentration occurring only after 5 hours of processing and dozens of passes of the model feed charge through the catalyst bed. These results simply cannot suggest that using higher liquid processing rates in the one-pass reactor of Soni would provide any improvement in the levels of sulfur removal achieved in the Soni process.

For the above reasons it is respectfully submitted that the combination of Soni and Irandoost cannot be read to suggest the processing conditions employed in the Applicants' hydrotreating process, nor do these references provide any reasonable expectation that high one-pass heteroatom removal levels could be achieved in such hydrotreating processes under the reaction conditions disclosed and claimed by the Applicants. Accordingly, reconsideration and withdrawal of the rejection of claims 1-15, 18-21 and 24 of the application on reference to these publications are respectfully requested.

Finally the Examiner has rejected claims 16, 17, 22, 23, 27-31 and 36-39 of the application under 35 U.S.C. §103 as unpatentable over Soni and Irandoost further in view of GB 936,941 (the '941 patent). The '941 patent was cited to show the use in a hydrotreating process of LHSV and gas:liquid ratio values within the scope of the Applicants' claims. This rejection is respectfully traversed for the following reasons.

The '941 patent relates to only novel catalyst formulations for more effectively removing nitrogen from hydrocarbon feedstocks. Moreover, the catalysts provided are pelletized catalysts and the reactors employed for processing are conventional trickle bed reactors, with no teaching or suggestion of monolithic catalysts and reactors as the Applicants' claims require.

As the Applicants and other prior art workers have recognized, conditions useful for the operation of trickle bed reactors are generally not applicable to the operation of monolith reactors. In particular, trickle bed reactors are generally operated at relatively low liquid linear flow velocities for best results, as the Applicants report at page 1, lines 21-28 of the specification:

Current commercial HDS technology is mature and is based on cobalt/molybdenum impregnated gamma-alumina or on nickel/molybdenum impregnated catalysts. These catalysts are employed in large reactors as random packed beds of spherical, cylindrical, or shaped extrudate beads. HDS reactors typically operate in a trickle-bed mode wherein the raw, high sulfur-containing distillate-range hydrocarbon liquid flows at relatively low velocity downward through the catalyst bed, while a hydrogen-rich treat gas flows co-currently downward through the catalyst at a much higher velocity.

Given this art-recognized distinction, to modify the monolith process of Soni by adopting the hydrogen:liquid ratios and higher space velocities of the '941 pellet bed process would not be an obvious step. This is particularly true since the Soni data clearly indicates reduced conversion efficiencies in monolith reactors at higher space velocities. Of course, even if the Soni process were modified in light of the '941 trickle bed conditions, the much higher liquid linear velocities of the Applicants' process would still not be taught or suggested. Nor is there anything in the '941 patent to suggest that liquid flow velocities higher than those conventional for trickle bed reactors should be used.

In summary, the processing conditions of Soni, Irandoost and the '941 patent, all having been established in light of the particular catalysts, reactor designs, and chemical processes described in each of the cases, cannot be arbitrarily selected and combined in the manner proposed by the Examiner without some suggestion in any of the references to combine the processing conditions in the manner proposed. Thus it appears that the combination of features now relied on by the Examiner in support of the rejection may be arrived at only in light of the Applicants' own disclosure. Clearly, this would constitute a hindsight reconstruction of the Applicants' invention that cannot serve as a proper basis for a rejection of the Applicants' claims.

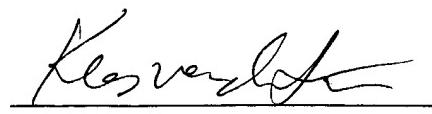
For these reasons, reconsideration and withdrawal of the rejection of claims 16, 17, 22, 23, 27-31 and 36-39 on reference to the combination of Soni, Irandoost, and the '941 patent are respectfully requested.

In light of the foregoing amendments and remarks, the Applicants respectfully submit that the remaining claims of this application are now in condition for allowance. Accordingly favorable reconsideration of this application and allowance of all of the remaining herein are courteously solicited.

Applicants believe that no extension of time is necessary to make this Reply timely, but contingently request that the Office grant such time extension pursuant to 37 C.F.R. § 1.136(a) as is necessary to make this Reply timely, if in fact such an extension is required. In that contingency the Office is hereby authorized to charge any necessary extension fee or surcharge to the deposit account of Corning Incorporated, Deposit Account 03-3325.

Respectfully submitted,

DATE: September 3, 2003



Kees van der Sterre  
Attorney for Assignee  
Reg. No. 25,938  
Corning Incorporated  
SP-TI-03-1  
Corning, NY 14831  
(607) 974-3294

**Amendments to the Claims:**

Please replace all prior claims versions and listings with the following:

1. (currently amended) A method for hydrotreating hydrocarbons to remove heteroatoms therefrom comprising:
  - a. combining hydrogen-containing gas with a liquid hydrocarbon stream containing sulfur, nitrogen, metal or oxygen heteroatoms to form a feed stream;
  - b. passing said feed stream over a monolithic catalyst bed containing hydrotreating catalyst components to remove the heteroatoms therefrom;
  - c. wherein the superficial liquid linear velocity of said feed stream is greater than about 0.2 cm/s, the feed hydrogen gas to liquid feed volume ratio is greater than about 50 NL/L, the liquid hourly space velocity is greater than about 0.7 h<sup>-1</sup>, the reactor pressure is greater than about 20 bar, the reaction temperature is greater than about 250°C 0.02 cm/s and the one-pass conversion of a heteroatom is greater than 50%.
2. (canceled)
3. (original) The method for hydrotreating hydrocarbons as described in claim 1, wherein said monolithic catalyst bed has a honeycomb configuration.
4. (canceled)
5. (original) The method for hydrotreating hydrocarbons as described in claim 1, wherein said hydrotreating catalyst components are from the group of cobalt, molybdenum, nickel, tungsten, and phosphorous.
6. (original) The method for hydrotreating hydrocarbons as described in claim 1, wherein said hydrotreating catalyst components are metals selected from Group VIII of the Periodic Table.

7. (original) The method for hydrotreating hydrocarbons as described in claim 1 wherein the monolithic catalyst bed comprises one or more inorganic honeycombs comprising channel wall surfaces formed of an alumina-containing catalyst support material.
8. (original) The method for hydrotreating hydrocarbons as described in claim 7 wherein the channel wall surfaces incorporate an alumina coating.
9. (original) The method for hydrotreating hydrocarbons as described in claim 7 wherein the inorganic honeycombs are formed of alumina.
10. (original) The method for hydrotreating hydrocarbons as described in claim 1 wherein the monolithic catalyst bed comprises one or more inorganic honeycombs comprising channel wall surfaces formed of a zeolite catalyst support material.
11. (original) The method for hydrotreating hydrocarbons as described in claim 1, wherein said liquid hydrocarbon stream has a boiling range within the range of about 70 to about 700°C.
12. (original) The method for hydrotreating hydrocarbons as described in claim 1, wherein the said liquid hydrocarbon stream is from a group of refinery streams consisting of distillates, gas oils, and gasoline blendstocks.
13. (original) The method for hydrotreating hydrocarbons as described in claim 1, wherein the said liquid hydrocarbon stream is in the diesel fuel boiling range.
14. (original) The method for hydrotreating hydrocarbons as described in claim 1, wherein the one-pass conversion of the targeted heteroatom is greater than 80%.
15. (original) The method for hydrotreating hydrocarbons as described in claim 1, wherein the one-pass conversion of the targeted heteroatom is greater than 90%.
16. (canceled)
17. (canceled)

18. (currently amended) A method for making low-sulfur diesel fuel comprising:
  - a. combining hydrogen-containing gas with a liquid hydrocarbon stream containing less than 3 wt % sulfur as organic sulfur compounds to form a feed stream;
  - b. passing said feed stream over a monolithic catalyst bed containing hydrotreating catalyst components at a superficial liquid linear velocity greater than about 0.2 cm/s, a feed hydrogen gas to liquid feed volume ratio greater than about 50 NL/L, a liquid hourly space velocity greater than about 0.7 h<sup>-1</sup>, a reactor pressure greater than about 20 bar, and a reaction temperature is greater than about 250°C to convert organic sulfur compounds to hydrogen sulfide and produce a treated hydrocarbon effluent;
  - c. separating the treated hydrocarbon effluent from a sour gas containing the hydrogen sulfide,
  - d. wherein the separated, treated liquid hydrocarbon is a diesel fuel contains less than about 5000 wppm sulfur.
19. (previously presented) The method for making low-sulfur diesel fuel as described in claim 18, wherein said diesel fuel product contains less than about 15 wppm sulfur.
20. (previously presented) The method for making low-sulfur diesel fuel as described in claim 18, wherein said monolithic catalyst bed has a honeycomb configuration.
21. (previously presented) The method for making low-sulfur diesel fuel as described in claim 18, wherein said hydrotreating catalyst components are from the group of cobalt, molybdenum, nickel, tungsten, and phosphorous.
22. (previously presented) The method for making low-sulfur diesel fuel as described in claim 18, wherein the superficial liquid linear velocity is greater than about 0.02 cm/s, the feed hydrogen gas to liquid feed volume ratio is greater than about 10 NL/L, the liquid hourly space velocity is greater than about 0.1 h<sup>-1</sup>, the reactor pressure is greater than about 1 bar, and the reaction temperature is greater than about 200°C.

23. (previously presented) The method for making low-sulfur diesel fuel as described in claim 22, wherein the feed hydrogen gas to liquid feed volume ratio is greater than about 50 NL/L, the liquid hourly space velocity is greater than about  $0.7\text{ h}^{-1}$ , the reactor pressure is greater than about 20 bar, and the reaction temperature is greater than about 250°C.

24. (previously presented) The method for making low-sulfur diesel fuel as described in claim 22, wherein the superficial liquid linear velocity is greater than about 0.2 cm/s.

25. (currently amended) A method for increasing one-pass heteroatom conversion in a hydrotreating reactor while maintaining hydrotreating selectivity, comprising:

- a. combining hydrogen-containing gas with a liquid hydrocarbon stream containing sulfur, nitrogen, metal or oxygen heteroatoms to form a feed stream;
- b. passing said feed stream over a monolithic honeycomb catalyst bed containing hydrotreating catalyst components to react the heteroatoms with hydrogen;
- c. said monolithic honeycomb catalyst bed having a cell density greater than about 10 cpsl and channel opening diameters greater than about 0.1 mm;
- d. wherein the superficial liquid linear velocity of said feed stream is greater than about 0.02 cm/s, the feed hydrogen gas to liquid feed volume ratio is greater than about ~~40-50~~ NL/L, the liquid hourly space velocity is greater than about ~~0.4-7~~  $\text{h}^{-1}$ , the reactor pressure is greater than about ~~±20~~ bar, and the reaction temperature is greater than about ~~200-250~~ °C;
- e. and wherein the one-pass conversion of the targeted heteroatom is greater than 50% with comparable product and feed distillation points.

26. (canceled).

| 27. (canceled)

| 28. (canceled)

29. (previously presented) The method for increasing one-pass heteroatom conversion in a hydrotreating reactor while maintaining hydrotreating selectivity as described in claim 25, wherein at least 70% (vol.) of the product stream has a D86 distillation temperature range falling within the D86 distillation range of the feedstock.

30. (previously presented) The method for increasing one-pass heteroatom conversion in a hydrotreating reactor while maintaining hydrotreating selectivity as described in claim 25, wherein said one-pass conversion of the targeted heteroatom is greater than 80%.

31. (previously presented) The method for increasing one-pass heteroatom conversion in a hydrotreating reactor while maintaining hydrotreating selectivity as described in claim 25, wherein said one-pass conversion of the targeted heteroatom is greater than 90%.

Claims 32-35. (withdrawn)

--36. (currently amended) A hydrotreating process for removing from a hydrocarbon feed stream in excess of 50% of the weight of sulfur present in that feed stream in a single pass through a monolith reactor, the feed stream having a boiling temperature range within the range of 70-700°C, which comprises the steps of:  
passing the feed stream and a hydrogen-containing gas through the reactor at a liquid superficial linear velocity in the range of about  $0.4\frac{m}{s}$  to about 10 cm/s, a hydrogen-containing gas-to-liquid volume ratio of about 10-2000 NL/L, a reaction temperature in the range of 200-500°C, a reactor pressures in the range of 100-2000 psig, and a liquid hourly space velocity (LHSV) in the range of  $0.4\frac{m^3}{m^3\cdot h}$  - 500  $\text{h}^{-1}$  to provide a hydrocarbon product stream. --

--37. (previously presented) A method in accordance with claim 36 wherein the hydrocarbon feed stream is a refinery or petrochemical liquid streams selected from the group consisting of petroleum distillates, gas oils, and gasoline blendstocks. --

--38. (previously presented) A method in accordance with claim 36 wherein the hydrocarbon product stream is a diesel fuel product containing less than 5000 wppm sulfur, and wherein at least 70% (vol.) of the diesel fuel product has a D86 distillation temperature range falling within the D86 distillation range of the hydrocarbon feed stream.--

--39. (currently amended) A method in accordance with claim 36 wherein in excess of 90% of the sulfur present in the feed stream is removed in a single pass through the reactor, and wherein: the liquid superficial linear velocity is in the range of 0.5 to about 5 cm/s, the hydrogen-containing gas-to-liquid ratio is in the range of 50-300 NL/L, the reaction temperature is in the range of 250 to about 400°C, the reactor pressure is in the range of 300-1000 psig., and the liquid hourly space velocity is in the range of 0.5<sup>7</sup>-50 h<sup>1</sup>--